

Spin Motion in Electron Transmission through Ultrathin Ferromagnetic Films Accessed by Photoelectron Spectroscopy

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(Dated: February 1, 2008)

Ab initio and model calculations demonstrate that the spin motion of electrons transmitted through ferromagnetic films can be analyzed in detail by means of angle- and spin-resolved core-level photoelectron spectroscopy. The spin motion appears as precession of the photoelectron spin polarization around and as relaxation towards the magnetization direction. In a systematic study for ultrathin Fe films on Pd(001) we elucidate its dependence on the Fe film thickness and on the Fe electronic structure. In addition to elastic and inelastic scattering, the effect of band gaps on the spin motion is addressed in particular.

PACS numbers: 75.70-i, 79.60.-i, 73.40.Gk, 75.50.Bb

To take advantage of the spin in electronic devices, in order to form new “spintronic” devices, is currently in progress worldwide. This goal challenges both applied and basic physics, the latter being mostly concerned with model systems of spin-dependent transport [1]. Aiming at very small devices, properties of magnetic nanostructures become increasingly important. In particular, spin-dependent scattering in ultrathin films and at interfaces may have a profound effect on the transport properties [2, 3]: the electronic spins start to precess and the spin current applies a spin-transfer torque on the magnetization in the ferromagnet. To understand in detail the spin motion in electron transmission through magnetic films, one obviously needs a microscopic probe.

Ferromagnetic resonance, used successfully to study magnetic properties of multilayer systems [4], unfortunately cannot deal with electron transmission. However, spin- and time-resolved photoelectron spectroscopy was employed to investigate directly spin filtering in the time domain [5]. Another successful method is transmission of spin-polarized electrons (usually produced with a GaAs source) through freestanding ferromagnetic films [6]. This way, the spin motion which shows up as precession of the electron spin-polarization (ESP) \vec{P} around the magnetization direction and as relaxation of \vec{P} towards the magnetization \vec{M} was investigated [7, 8].

In this Letter, we propose to apply angle- and spin-resolved photoelectron spectroscopy from core levels to access directly the spin motion of electrons transmitted through an ultrathin ferromagnetic film (Fig. 1). The “theoretical experiments” reported here rely in particular on the possibility to orient the spin polarization of the incoming photoelectrons by the incident light, an effect which is due to spin-orbit coupling. In the following, the basic ideas of our approach are described for the chosen systems n ML Fe/Pd(001), $n = 1, \dots, 6$ (for details, see Ref. [9]).

(i) The incident light excites electrons from Pd- $3d_{3/2}$ core levels of the Pd(001) substrate into a state above the vacuum level E_{vac} .

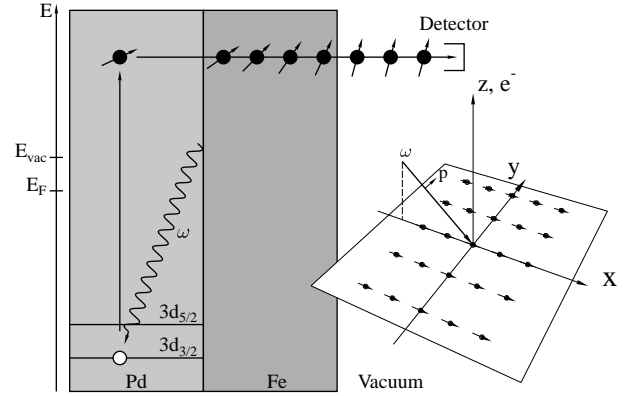


FIG. 1: Spin motion in electron transmission through a ferromagnetic film accessed by photoelectron spectroscopy. Left: A core electron is excited by the incident radiation (wavy line, photon energy ω) in the Pd substrate (light grey). The spin polarization (arrow) of the photoelectron (filled circle) is oriented due to spin-orbit interaction. During the transmission through the magnetic Fe film (dark grey), the spin polarization rotates further (spin motion) but stops rotating in the vacuum. E_F and E_{vac} are the Fermi and the vacuum level, respectively. Right: Set-up of normal photoemission from a ferromagnetic surface (with magnetization along x) and p-polarized light incident in the xz plane (photon energy ω).

(ii) Choosing linearly p-polarized light with incidence direction given by $\vartheta_{ph} = 45^\circ$ polar angle and variable azimuth φ_{ph} , the photoelectron spin in the substrate can be aligned to any desired direction in the xy surface plane (Cartesian coordinates are defined in Fig. 1). It was theoretically and experimentally shown for non-magnetic layered systems with fourfold rotational symmetry that an ESP perpendicular to the scattering plane (spanned by the surface normal and the incidence direction; see Ref. [10] and refs. therein) is produced: $\vec{P}^{in} \propto (-\sin \varphi_{ph}, \cos \varphi_{ph}, 0)$. For $\varphi_{ph} = 0^\circ$ and 180° , \vec{P}^{in} is perpendicular to the magnetization \vec{M} (which is parallel to x). Hence, the commonly used external GaAs

source for spin-polarized electrons is, so to speak, replaced by an internal one, with the advantage of easy orientation of \vec{P}^{in} . Choosing other light polarizations and incidence directions one can produce \vec{P}^{in} with a component along the surface normal [11].

(iii) During the transmission through the Fe film, the photoelectron is subject to elastic and inelastic processes. Both can simply be modeled by spin-dependent scattering at an asymmetric quantum well which comprises the substrate-film and the film-vacuum interfaces. The transmitted ESP \vec{P}^{tr} reads

$$\vec{P}^{\text{tr}} \propto \begin{pmatrix} |T^\uparrow|^2 - |T^\downarrow|^2 + P_x^{\text{in}}(|T^\uparrow|^2 + |T^\downarrow|^2) \\ P_y^{\text{in}}\text{Re}(T^{\uparrow*}T^\downarrow) - P_z^{\text{in}}\text{Im}(T^{\uparrow*}T^\downarrow) \\ P_z^{\text{in}}\text{Re}(T^{\uparrow*}T^\downarrow) + P_y^{\text{in}}\text{Im}(T^{\uparrow*}T^\downarrow) \end{pmatrix}, \quad (1)$$

where the spin-dependent transmission coefficients $T^{\uparrow(\downarrow)}$ take into account multiple scattering. Considering elastic scattering only, the dependence of \vec{P}^{tr} on the film thickness d shows two typical oscillation periods that depend on the electron wavenumbers $k_z^{\uparrow(\downarrow)}$ in the film. The longer period with wavelength $2\pi/(k_z^\uparrow - k_z^\downarrow)$ describes the precession of the transversal components P_y^{tr} and P_z^{tr} around \vec{M} [12, 13]. The short-period oscillation with wavelength $2\pi/(k_z^\uparrow + k_z^\downarrow)$ and much smaller amplitude is due to multiple scattering at the interfaces. The longitudinal component P_x^{tr} remains constant on average.

Inelastic scattering leads to spin-dependent attenuation within the film. This can simply be simulated by multiplying the phase factors that describe the propagation between the interfaces by $\exp(-d/\lambda^{\uparrow(\downarrow)})$. This spin-filter effect relaxes \vec{P}^{tr} towards \vec{M} (i.e., $\lim_{d \rightarrow \infty} P_x^{\text{tr}} = 1$ for $\lambda^\uparrow > \lambda^\downarrow$). It was successfully used to determine the attenuation lengths $\lambda^{\uparrow(\downarrow)}$ [14, 15] and to obtain the spin-resolved electronic structure of Fe [16]. There is no spin motion in nonmagnetic regions (e.g., vacuum).

(iv) The photoelectrons are eventually detected spin-resolved in normal emission ($\vec{k}_\parallel = 0$). Note that the electron energies are considerably larger than those in spin-dependent transport measurements.

The small photoelectron escape depth [17, 18] restricts d to a few ML. This implies for ultrathin films that the short-period oscillation might dominate the spin motion, a complete precession cannot be observed, and the relaxation limit ($\vec{P} \parallel \vec{M}$) cannot be reached in practice.

Theoretical. Starting from first-principles electronic-structure calculations for 0–6 ML fcc-Fe/Pd(001) (local spin-density approximation of density-functional theory; screened KKR method; for details, see Ref. [9]) we computed spin- and angle-resolved constant-initial state photoemission spectra within the relativistic one-step model, as formulated in the layer-KKR method (see, e.g., Ref. [19]).

We choose Fe/Pd(001) due to the large magnetic moment of Fe and the strong spin-orbit coupling in Pd which results in a sizable \vec{P}^{in} . The covering Fe induces a mag-

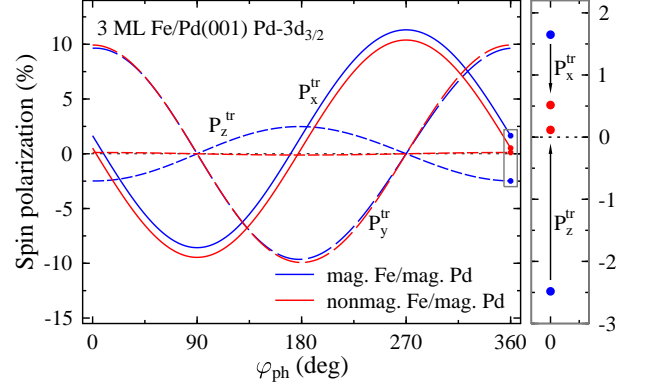


FIG. 2: Transmitted spin polarization \vec{P}^{tr} versus azimuth φ_{ph} of light incidence for 3 ML Fe/Pd(001) and 27 eV kinetic energy. Blue (red) lines are for magnetic (nonmagnetic) Fe on magnetic Pd (see text). The right panel shows data for $\varphi_{\text{ph}} = 0^\circ$ or 360° enlarged (cf. the grey rectangle).

netic moment of about $0.24 \mu_B$ in the Pd layer close to the Fe/Pd interface [9]. Hence, \vec{P}^{in} originates from the induced exchange splitting and from spin-orbit coupling. To reveal the origin of the spin motion, we considered various “artificial” magnetic configurations in which the magnetization in the Fe film or in the Pd substrate were switched off separately. Further, changing the inverse photoelectron lifetime in the Fe film allows to differentiate between elastic (precession around \vec{M}) and inelastic processes (relaxation towards \vec{M}).

Existence and origin of the spin motion. Considering the configuration with all Fe layers and the Pd layers close to the Fe/Pd interface being magnetic (“mag. Fe/mag. Pd”, blue in Fig. 2) and \vec{P}^{in} along the y axis ($\varphi_{\text{ph}} = 0^\circ$; right panel), the existence of the spin motion is established by nonzero P_x^{tr} and P_z^{tr} . For $\varphi_{\text{ph}} = 90^\circ$ and 270° (i.e., $\vec{P}^{\text{in}} \parallel \vec{M}$) both P_y^{tr} and P_z^{tr} vanish, leaving only P_x^{tr} nonzero in agreement with Eq. (1). The dependence of \vec{P}^{tr} on φ_{ph} (left panel) follows perfectly that derived analytically in Ref. [11].

Setting all Fe layers artificially nonmagnetic but keeping the magnetism in the Pd interface layers (“nonmag. Fe/mag. Pd”, red in Fig. 2), P_z^{tr} is strongly reduced (in absolute value). This confirms that the spin motion originates dominantly from the magnetism in the Fe film. In the complete nonmagnetic configuration (not shown), P_z^{tr} vanishes.

Elastic and inelastic processes. Inelastic processes can be simulated in calculations by adding an imaginary self-energy to the potential (see, e.g., Ref. [20]). To unveil the influence of these processes, we reduced the inverse photoelectron lifetime in the Fe film to 0.001 eV (“elastic” case), compared to the otherwise chosen 1.8 eV (“inelastic” case). Being rather small and almost constant in the “elastic” case, P_x^{tr} increases with Fe coverage

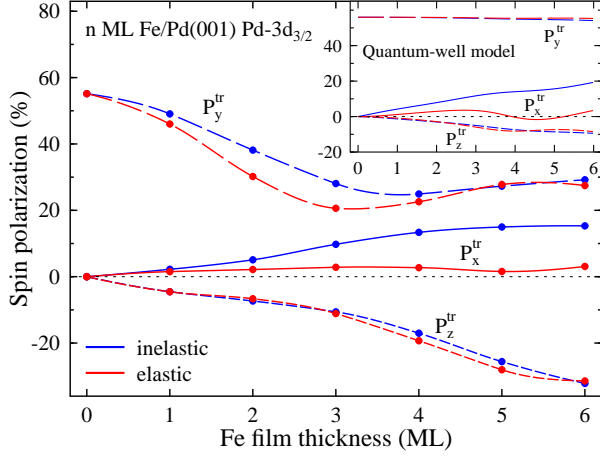


FIG. 3: Elastic and inelastic effects in electron transmission through 0–6 ML Fe on Pd(001) at 17.5 eV kinetic energy and azimuth of light incidence $\varphi_{\text{ph}} = 0^\circ$. The transmitted electron-spin polarization \vec{P}^{tr} is shown versus Fe-film thickness n (in ML) for the “inelastic” (blue) and the “elastic” (red) case. The inset shows corresponding results of a model calculation.

in the “inelastic” case (Fig. 3), i.e., \vec{P}^{tr} starts to relax towards \vec{M} . Because the short-period oscillation is relevant for ultra-thin films, the precession of \vec{P}^{tr} around \vec{M} (which shows the long wavelength) cannot be clearly observed. To corroborate these findings, we calculated \vec{P}^{tr} within the quantum-well model sketched preceding, with parameters obtained from the Pd and Fe bulk-band structures (inset in Fig. 3). The resulting wavelengths of about 200 ML (precession) and 3.9 ML (multiple-scattering) lead to reasonable agreement concerning P_x^{tr} and P_z^{tr} . However, P_y^{tr} does not show such pronounced a minimum at 3–4 ML. The differences between model and *ab initio* calculations can be attributed to the number of transmission channels: a single one in the model but several channels (with different wavelengths) in the *ab initio* calculations.

Effects of the electronic structure. To show how the spin motion depends on details of the electronic structure, we address spin-resolved constant-initial-state photoemission spectra. In contrast to SPLEED experiments in which \vec{P}^{in} is typically parallel or antiparallel to the magnetization [21, 22], we choose a transverse \vec{P}^{in} ($\varphi_{\text{ph}} = 0^\circ$). For clarity reasons, the following discussion rests upon the complex bulk band structure, rather than on layer- and spin-resolved spectral densities.

The spin-averaged intensities (Fig. 4a) decrease significantly with Fe coverage, caused by the small photoelectron escape depth. The global shape of the spectra, however, remains almost unaffected. Changes of the slopes, best to be seen for 1 ML Fe but present for all Fe-film thicknesses, can be traced back to the Fe elec-

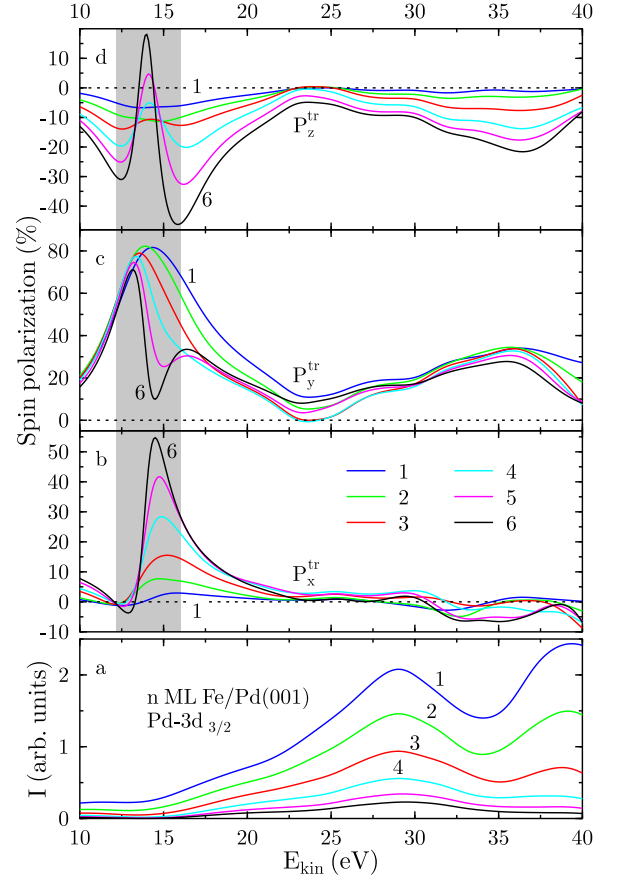


FIG. 4: Energy dependence of the spin motion for 1–6 ML Fe on Pd(001). (a) Spin-averaged constant-initial-state photoemission intensities I versus kinetic energy E_{kin} of the photoelectrons. (b)–(d) Transmitted electron-spin polarization \vec{P}^{tr} . The Fe-film thickness n (in ML) is indicated by numbers and color-coding. The grey area highlights a prominent feature discussed in the text.

tronic structure (not shown): an increase of the slope is associated with the onset of additional transmission channels, i.e., dispersive Fe bands. In particular, one pair of spin-split bands provides efficient transmission channels, which leads to the intensity increase at about 15 eV. The pronounced minimum at about 34 eV kinetic energy can be attributed to a Pd-band gap, which reduces the number of transmission channels in the substrate.

At higher energies where several transmission channels contribute, the evolution of \vec{P}^{tr} with Fe coverage is rather complicated (Figs. 4b–d). Therefore, we concentrate on low energies where the number of channels is small and the evolution is almost monotonous. The most significant structures that increase with Fe coverage show up between 12 eV and 16 eV (grey area in Figs. 4b–d): P_x^{tr} and P_y^{tr} display $-/+$ and $+/-$ modulations, resp., accompanied by a maximum in P_z^{tr} . A detailed analysis corroborates its relation to the Fe electronic structure

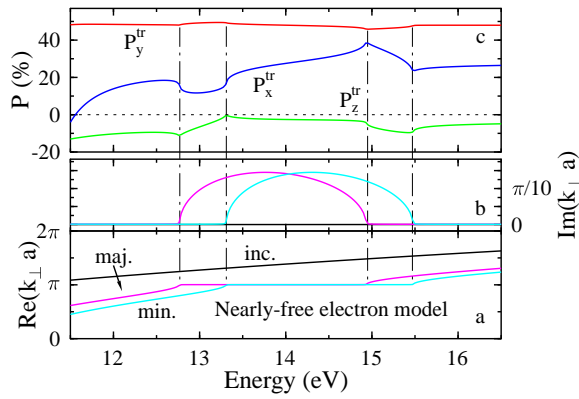


FIG. 5: Effect of exchange-split band gaps in the film electronic structure on the spin motion. (a) and (b): Complex band structure of the substrate (dash-dotted, “inc.”) and the magnetic film (majority, solid, “maj.”; minority, dashed, “min.”) in the extended zone scheme. (c) Electron spin polarization of the transmitted electrons. For details, see text. Vertical dash-dotted lines serve as guides to the eye.

and shows that it can be attributed to exchange-split band gaps in conjunction with the onset of additional transmission channels in that particular energy range.

To provide direct evidence that band gaps manifest themselves in pronounced spin-motion structures, we calculated the ESP in an inelastic three-band nearly-free-electron model. The substrate is taken as semi-infinite free space (with zero potential), whereas a nonzero scattering potential in the magnetic film gives rise to exchange-split band gaps (Figs. 5a and b). For $P_y^{\text{in}} = 50\%$ in a representative set-up, P_x^{tr} and P_y^{tr} show a $-/+$ and a small $+/-$ modulation, resp., whereas P_z^{tr} increases in the band-gap middle (Fig. 5c). These findings can be explained by the reduced transmission of one spin channel which is mediated by evanescent states [nonzero $\text{Im}(k_z)$ in Fig. 5b; note that transverse spinors are given by a weighted sum of spin-up (“maj.”) and spin-down (“min.”) Pauli spinors]. Although the Fe band structure is much more complicated, the structures in the model calculation have counterparts in Figs. 4b–d (grey area). That distinct band-gap related features do not show up in Fig. 4 at higher kinetic energies just at about 15 eV.

Conclusions. We have shown by means of “theoretical experiments” that the spin motion in electron transmission through ferromagnetic films can be analyzed in detail by angle- and spin-resolved photoelectron spectroscopy. As main advantages of this approach one might consider that the preparation of freestanding films is avoided and that the spin polarization of the incoming electrons can easily be oriented. Since intensities and spin polarizations depend significantly on the film thickness, one ob-

tains information on the electronic structure, in particular on that of the film. Realistic calculations for Fe films on Pd(001), that are to be confirmed experimentally, suggest promising analyses of spin-dependent transport through magnetic layers. Further, we regard our approach as a tool for investigations of magnetic configurations, with the possibility of analyzing noncollinear spin structures.

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